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# Contactless conductivity measurements on $La_2CuO_4$ at radio frequencies

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The experimental setup for contactless conductivity measurements is reported which can easily be operated at frequencies $10^6 \leq \nu \leq 10^9$ Hz. The technique is applied to complex conductivity measurements in $La_2CuO_4$ at temperatures from 4 to 300 K. The results are compared with data obtained by conventional techniques.

## I. INTRODUCTION

The dielectric properties of $La_2CuO_4$ are still not well understood. The complex permittivity has been studied in the microwave regime[1] and at audio and radio frequencies.[2,3] The results of these studies are not fully consistent and the interpretations differ drastically. The microwave results were interpreted assuming an unusual large effective mass, while the results in the audio and radio frequency regime revealed a behavior typical for conventional semiconductors.[2] In addition, the ac conductivity is characterized by a power-law dependence on frequency. But the temperature dependence of the frequency exponent is very different in different experimental setups.[2,3] We thought that these discrepancies are partly due to problems of contact resistances for the frequencies below 15 GHz.[1-3] In this report we describe an experimental setup for contactless conductivity measurements and we show some results of the complex conductivity in $La_2CuO_4$ at temperatures from 4 to 300 K. However, we also would like to point out, that the same method can also be used for a variety of semiconducting and ionic conducting materials.

Measuring techniques for the ac conductivity in the audio and subaudio frequency range ($10^0 < \nu < 10^6$ Hz) are well established and commonly applied. At these frequencies standard *LCR* meters or two-phase lock-in amplifier techniques can be used. Depending on the order of magnitude of the complex conductivity, four-probe or two-probe configurations are adapted. Again, dedicated measuring techniques have been developed at microwave frequencies ($10^9 < \nu < 10^{11}$ Hz) utilizing wave guides and cavities. In between these two frequency regimes commercial network analyzers can be operated from 1 MHz to 10 GHz and the radiowave transmission is managed using unmatched air lines or special coaxial cables. But for these techniques two-probe configurations have to be used and the measured conductivities have to be analyzed using electronic equivalent circuits in order to distinguish between intrinsic properties of the sample and the unwanted influence of the contacts. In semiconducting materials the contact resistance can be by orders of magnitude larger than the sample resistance, thus hampering a reliable estimate of the intrinsic conduction properties.

A further problem is that the rf measuring equipment has to be calibrated with 0 Ω (short circuit), a load resistance (typically 50 Ω), and 0 S (open circuit). Calibration procedures can readily be performed at room temperature, but are extremely time consuming when the temperature-dependent conductivities are measured in a wide temperature range using cryostat or oven equipment (e.g., a precise 50-Ω resistor has to be designed for every temperature at which the sample should be investigated).

A set of alternative methods has been presented in the literature, allowing one to carry out contact-free measurements. The proposed methods use, instead of direct electric contacts, capacitive[4,5] or inductive[6] coupling of the rf waves to the sample. In the following we describe a method to measure the ac conductivity in the radio frequency range ($1 < \nu < 500$ MHz) utilizing the idea of a capacitive coupling of the sample to a resonant circuit, similar to the proposal of Miyamoto and Nishizawa.[4] However, we show that it is feasible to measure both the shift of the resonance frequency $\Delta\nu_0$ and the change of the quality factor $\Delta Q$ using an extremely simple experimental setup. Furthermore we demonstrate that measurements can easily be performed at cryogenic and at elevated temperatures. This is due to the fact that the simultaneous determination of $\Delta\nu_0$ and $\Delta Q$ allows one to avoid the initial calibration procedures which have to be performed using the methods proposed in Refs. 4 and 5. Our technique is most appropriate for the investigation of samples which are characterized by hopping conductivity processes, which are dominant in, e.g., amorphous semiconductors or ionic conductors. In this report, we present the results of contactless measurements on $La_2CuO_4$ for frequencies $3 < \nu < 300$ MHz and temperatures $4 < T < 300$ K and compare them to results as obtained by conventional techniques.

## II. EXPERIMENTAL DETAILS

The complex conductivity is obtained by measuring the resonance frequency $\nu_0$ and the quality factor $Q$ of a *LC* circuit at resonance conditions with and without sample. A schematic illustration of the measuring device is depicted in the upper frame of Fig. 1. We have successfully investigated different semiconducting samples with typical sizes of a few $mm^3$. Of course, samples with plane parallel faces give superior results but are not necessarily needed. The electronic equivalent circuit of the measur-

 

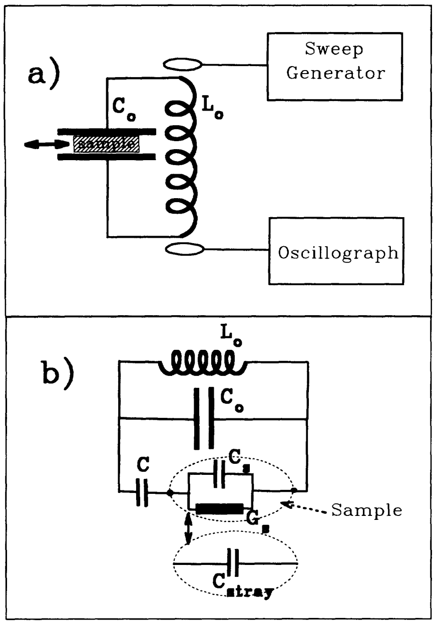


FIG. 1. (a) Schematic illustration of the measuring device. (b) Electronic equivalent circuit of the experimental setup. $L_0$ and $C_0$ are the inductance and the capacitance of the resonant circuit. $C_s$ and $G_s$ are capacitance and conductance of the sample. $C$ is the coupling capacitance and $C_{\text{stray}}$ the stray capacitance.

ing device could be represented as shown in the lower panel of Fig. 1. $L_0$, $C_0$, and $G_0$ are the inductance, capacitance and equivalent conductance of the resonance circuit. $C_s$ and $G_s$ are the capacitance and the conductance of the sample. $C$ is the coupling capacitance which depends on the geometries of the sample and of the empty capacitor. Finally, $C_{\text{stray}}$ denotes the "empty" sample and can be determined experimentally. Typical values of the components as shown in the equivalent circuit are $L_0=10^{-5}$ H, $C_0=10$ pF, $C_s=1$ pF, $1/G_s=10$ kΩ, $C=0.1$ pF, and $C_{\text{stray}}=0.01$ pF.

Within the approximations of the used equivalent circuit, the change in the resonance frequency $\Delta\omega$ and in the inverse quality factor $\Delta(Q^{-1})$ are calculated according to

$$\Delta\omega/\omega_0=C/2C_0\ , \tag{1}$$

$$\Delta(Q^{-1})=\omega^3C^2L_0R_s\ . \tag{2}$$

The real part of the sample resistance $R_s$ is given by

$$R_s=G_s/(G_s^2+\omega^2C_s^2)\ . \tag{3}$$

Here we used the angular frequency $\omega=2\pi\nu$.

Note, that the intrinsic losses of the resonant circuit do not enter in Eqs. (1) and (2). In Eqs. (1) and (2) it is assumed that in an experiment the geometries of the sample and the capacitance $C_0$ are chosen in such a way that the coupling capacitance $C \ll C_s$. All the calculations could be done without this assumption, they only become in that case, slightly more complicated. The value of the coupling capacitance from Eq. (1) can be used to determine the conductivity of the sample via Eq. (2) despite the fact that this equation still contains the two unknown sample parameters $C_s$ and $G_s$. This will be demonstrated in the following.

If the sample under investigation is mainly conducting, i.e., $G_s \gg \omega C_s$ it follows immediately from Eq. (3) that $R_s=1/G_s$. However, even under the condition that the capacitive resistance dominates, both quantities, viz. $G_s$ and $C_s$, can be extracted from Eq. (3) if the sample capacitance $C_s$ is only weakly temperature dependent. Under these assumptions, $R_s$ as a function of $G_s$, and correspondingly as a function of $T$, will reveal a maximum when $1/R_s=G_s/2=\omega C_s/2$. Hence $G_s$ and $C_s$ can be calculated for all temperatures.

In the following we want to show that these constraints are almost perfectly met in many samples characterized by hopping conductivity. Hence the conductivity in these samples can be measured in a broad temperature range. In samples characterized by hopping conduction processes, the sample conductivity $G_s$ and the sample capacitance $C_s$ are given by[7]

$$\begin{aligned} G_s&=C_g(\sigma_{\text{dc}}+A'\omega^s)/\varepsilon_0\ ,\\ C_s&=C_g(\varepsilon_\infty+A''\omega^{s-1}/\varepsilon_0)\ . \end{aligned} \tag{4}$$

$\varepsilon_0$ is the permittivity of free space. $C_g$ is the geometrical capacitance.

In amorphous semiconductors and ion conductors the dc conductivity $\sigma_{\text{dc}}$ follows an exponential temperature dependence. The real part of the ac conductivity $A'\omega^s$ is characterized by a much weaker $T$ dependence and by a power law in the frequency domain with $s<1$.[7] The real and imaginary parts of the ac conductivity are related via the Kramers-Kronig relation and usually reveal similar dependencies on temperature and frequency. $\varepsilon_\infty$ is the high-frequency contribution to the dielectric constant and is only weakly temperature dependent.

At high temperatures $\sigma_{\text{dc}}$ dominates all contributions to the hopping conductivity. Hence, $G_s \gg \omega C_s$ and $R_s \sim 1/G_s$. At low $T$, the dc and ac contributions become small and the leading term will be $C_s\varepsilon_\infty$. Thus, at low temperature $\omega C_s$ dominates corresponding to our constraint that $\omega C_s \gg G_s$. However, it is important to note that $G_s$ should only weakly depend on $T$.

## III. EXPERIMENTAL RESULTS AND DISCUSSION

The aim was to study with this experimental technique $La_2CuO_4$ single crystals at different measuring frequencies for temperatures $4 \le T \le 300$ K. The results of the resistance with the field in the $c$ direction are shown in Fig. 2. With decreasing $T$, the resistance increases for all frequencies, passes through a maximum, and decreases for further decreasing temperatures. As outlined above, the maximum resistance allows for an unambiguous determination of the real *and* the imaginary parts of the conductivity.

At the temperature of the maximum resistance $\sigma_{\text{dc}} \approx \varepsilon_0\omega\varepsilon_\infty$. Hence the maximum resistance should reveal the temperature dependence of the dc conductivity which, in the case of $La_2CuO_4$ can be described by the

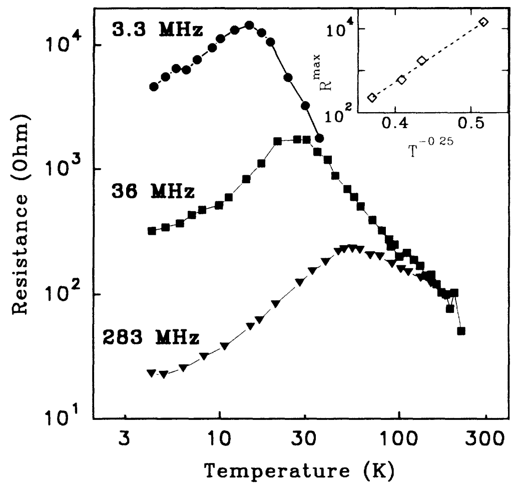


FIG. 2. Temperature dependence of the resistance in $La_2CuO_4$ as measured with the field parallel to the $c$ axis at three frequencies, shown in a double-logarithmic representation. The inset demonstrates that the dc conductivity, as obtained from $R_{max}$, follows Mott's variable-range-hopping law.

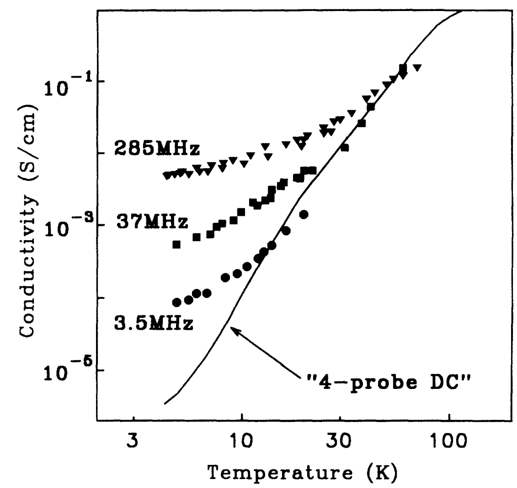


FIG. 4. Conductivity $\sigma'$ in $La_2CuO_4$ for fields within the $ab$ plane vs temperature for different measuring frequencies: ▼ : 285 MHz, ■: 37 MHz, ⊙: 3.5 MHz. Also indicated are the results of a four-probe dc measurement (solid line, Ref. 8), taken on the same single crystal.

Mott variable-range-hopping model. The result is shown in the inset of Fig. 2 where we plotted the logarithm of $R_{max}$ vs $T^{-1/4}$. The straight line indicates that our results follow the expected behavior. Using Eq. (3) we now proceed to calculate $G_s$, the real part of the conductivity. The results are shown in Figs. 3 and 4 in comparison with conventionally measured data. Figure 3 reveals the conductivity in $La_2CuO_4$ for the fields along $c$, Fig. 4 for fields within the $ab$ plane. The results of the contactless measurements are compared with the four-probe dc results. For $T > 50$ K all data fall onto a common line, the signature of the pure dc conductivity. In this high-temperature regime the contactless data fit well to the four-probe dc conductivity data measured on the same single crystal.[8] At low $T$ the ac conductivity dominates, indicating a power-law behavior according to $A\omega^s$. For temperatures below 15 K it was possible to estimate the value of frequency exponent $s$. The results are shown in Fig. 5. Despite the fact that the experimental errors are rather large, the different frequency exponents for the two directions can clearly be seen. Both frequency exponents $s$ decrease with increasing temperature. However these dependencies are much weaker, than those, reported in Ref. 2, where $s$ as measured in the $ab$ plane decreases between 4 and 12 K from $s = 0.77$ to $s = 0.51$. We believe, that this strong temperature dependence, as well as the small values of $s$ for the $c$ direction ($s \approx 0.25$, Ref. 3) are probably due to undesirable influences of contact resistances.

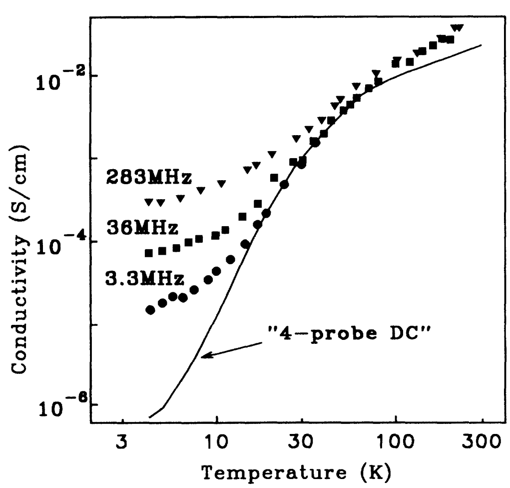


FIG. 3. Conductivity $\sigma'$ in $La_2CuO_4$ along the $c$ axis vs temperature for different measuring frequencies: ▼ : 283 MHz, ■: MHz, ⊙: 3.3 MHz. Also indicated are the results of a four-probe dc measurement (solid line, Ref. 8), taken on the same single crystal.

From the maximum resistance in Fig. 2, $\varepsilon'$ for the $c$ axis was determined to be $29 \pm 4$ independent of the measuring frequency. This result coincides with that

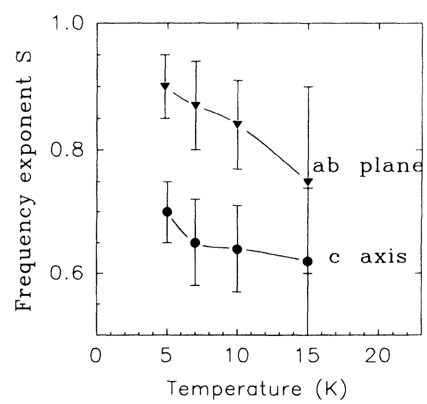


FIG. 5. Frequency exponent $s$ in $La_2CuO_4$ according to a power-law behavior vs temperature ($\sigma = \sigma_0 + A\nu^s$). The values are calculated on the basis of data presented in Figs. 3 and 4.

from two-probe measurement in the same single crystal which yielded $\varepsilon'=31\pm3$.[8] Within the *ab* plane we found $\varepsilon'=90\pm10$, which, in the single crystal investigated, could not be directly compared with the contact data because of a much stronger influence of contacts and residual ac components. Typical two point values[8] lie in the range from 60 to 110.

## IV. CONCLUSIONS

In this report we have presented a simple contactless method which allows one to determine the ac conductivity in semiconductors at radio frequencies. We outlined the assumptions necessary for an unambiguous calculation of the complex conductivity. The method yields reliable results for (i) conducting samples with $G_s \gg \omega C_s$ or (ii) for samples with a strongly temperature-dependent conductivity $G_s$ but with a temperature-independent capacitance $C_s$. We have shown that these constraints are almost perfectly met in amorphous semiconductors and ionic conductors which are characterized by hopping conductivities. The method is applied to the measurements of the ac conductivity in $La_2CuO_4$ single crystals at frequencies ranging from 3 to 300 MHz and at temperatures from 4 to 300 K. A comparison with results obtained using a four-probe geometry demonstrates the reliability of the method. In the present investigation the results obtained by these contactless measuring techniques seem to be superior compared to the results from two-point configuration and, in any case, they provide an important check of the reliability of conductivity measurements using conventional techniques.

## ACKNOWLEDGMENTS

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